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THE EFFECT OF CYCLIC PULSED TEMPERATURE ON VOID GROWTH IN METALS DURING IRRADIATION*

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The analysis of Brailsford and Bullough on radiation-produced point defect transport to lattice sinks and the analysis of Ghoniem and Kulcinski of pulsed radiation have been extended to include the effects of radiation-induced temperature oscillations. We have already shown in earlier papers that these temperature pulses enhance void growth if the ambient temperature is below the peak swelling temperature and, conversely, decrease void growth when the ambient temperature is above the peak swelling temperature. When the temperature pulses were suppressed, no difference was seen between void growth under steady and pulsed irradiation. In this paper, we show that the same effects on void growth can be obtained by superposing externally-produced temperature pulses on material under steady irradiation. Calculations have been done for aluminum and molybdenum for sinusoidal temperature pulses of many amplitudes and frequencies.

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1. INTRODUCTION

The fully dynamic rate theory, developed for steady state conditions by Brailsford and Bullough [1-3], was used by Ghoniem and Kulcinski [4,5] to calculate void growth and swelling for a single radiation pulse impinging on a constant ambient temperature material. This theory has been extended to follow the effects of arbitrary numbers of repeated pulses and their associated radiation-produced temperature oscillations on material damage structure [6,7].

Numerical calculations for aluminum and molybdenum showed that the time dependence of such self-consistent temperature changes must be taken into account because the amount of void growth is a sensitive function of temperature. Pulsing the irradiation had only an indirect effect on material swelling. When the cyclic radiation was allowed to produce associated temperature pulses, the material saw enhanced ambient temperatures. This temperature enhancement increased void growth if the ambient temperature was below the material peak swelling temperature, and conversely decreased void growth when the ambient temperature was above peak swelling temperature. (The exception seen occurred when the temperature pulse was great enough that, for some period of time, thermal emission of vacancies dominated radiation production.) When radiation-produced temperature pulses were suppressed so that the material remained at a constant ambient temperature, the void growth did not change as the radiation was fragmented into various pulse structures.

The present work continues the study of the indirect effect of pulsed radiation on material swelling by imposing externally-produced sinusoidal temperature pulses on materials under-

going steady irradiation. This may provide a way to simulate the effects of pulsed irradiation using more commonly available steady state devices. Swelling rates in aluminum and molybdenum are calculated for conditions similar to those used in the pulsed radiation studies, and the effect on void growth of varying amplitude and frequency of the applied sinusoidal temperature pulses is investigated.

These calculations form the design baseline for an irradiation experiment at the Clinton P. Anderson Los Alamos Meson Physics Facility in which identical parameters will be set to allow comparison of theory and experiment. The result will be a self-consistent determination of material response to conditions that simulate the environment for structural and confinement materials in fusion reactors.

2. METHOD

The six coupled simultaneous nonlinear ordinary differential equations solved in the fully dynamic theory are:

$$\dot{r}_v = \frac{1}{r_v} \left\{ D_v C_v - D_i C_i - D_v C_v^0 \exp \left[\left(\frac{2v}{r_v} - p_g \right) \frac{b^3}{kT} \right] \right\} \quad (1)$$

$$\dot{r}_{v1} = \frac{1}{b} \left\{ z_1 D_i C_i - z_v D_v C_v + z_v D_v C_v^0 \exp \left[\frac{-(1v + p_g) b^3}{kT} \right] \right\} \quad (2)$$

$$\dot{N}_{v1} = \frac{cP}{ab[r_{v1}(0)]^2} + \frac{N_{v1}}{br_{v1}(0)} \left\{ z_v D_v C_v - z_1 D_i C_i \right\}$$

$$\frac{-N_{vI}}{b r_{vI}(0)} \left\{ Z_v D_v C_v^0 \exp \left[\frac{(Y_{sf} + F_{el}) (r_{vI}(0))^2}{kT} \right] \right\} \quad (3)$$

$$\dot{r}_{vI} = \frac{1}{b} \left\{ Z_v D_v C_v - Z_i D_i C_i - Z_v C_v D_v \exp \left[\frac{+(Y_{sf} + F_{el}) b^2}{kT} \right] \right\} \quad (4a)$$

$$\dot{q}_v = c_p - 2\pi L r_{vI} N_{vI} \dot{r}_{vI} \quad (4b)$$

$$\dot{C}_i = P - a C_i C_v$$

$$-C_i D_i \left\{ 4\pi r_{vI} N_{vI} + Z_i \left[C_d^0 + 2\pi (r_{iI} N_{iI} + r_{vI} N_{vI}) \right] \right\} \quad (5)$$

$$\dot{C}_v = (1-c)P - a C_i C_v$$

$$-C_v D_v \left\{ 4\pi r_{vI} N_{vI} + Z_v \left[C_d^0 + 2\pi (r_{iI} N_{iI} + r_{vI} N_{vI}) \right] \right\}$$

$$+ 4\pi r_{vI} N_{vI} C_v \exp \left\{ \left(\frac{2Y}{r_v} - P_g \right) \frac{b^3}{kT} \right\}$$

$$-Z_v C_v D_v \left\{ C_d^0 + 2\pi \left[r_{iI} N_{iI} \exp \left(\frac{-(Y_{sf} + F_{el}) b^2}{kT} \right) \right] \right\}$$

$$+ r_{vI} N_{vI} \exp \left\{ \frac{+(Y_{sf} + F_{el}) b^2}{kT} \right\} \quad (6)$$

where C_v is the vacancy concentration, C_i is the interstitial concentration, r_v is the void radius, r_{iI} is the interstitial loop radius, q_{vI} is the fraction of vacancies in vacancy loops and r_{vI} is the vacancy loop radius. All other symbols have their usual meaning, as in the work of Ghoniem and Kulcinski as well as Brailsford and Bullough.

The rate equations include radiation point defect production, recombination and absorption, as well as thermal emission from all sinks. An edge dislocation network is included and is held at a constant density during absorption and emission of point defects. An oscillating time-dependent temperature is used in the appropriate equations as well as in the expressions for the diffusion coefficients. D_i and D_v

$$D_{i,v} = D_{i,v}^0 \exp \left(\frac{-E_{i,v}}{kT} \right) \quad (7)$$

(These temperature oscillations could also produce fluctuating thermoelastic and thermoplastic stresses, which are not treated in the present work.) The temperature is assumed to be given by

$$T(t) = T_0 + T_A \sin \left(\frac{t}{\tau} \right) \quad (8)$$

Damage energy cross sections derived by Collier (8.9) were used in the displacement damage calculations.

TABLE 1
Assumptions and Constants

Dislocation Density, ρ_d^0	10^{11}	m^{-2}
Void Number Density, N_v	10^{20}	m^{-3}
Interstitial Loop Number Density, N_{iI}	10^2	m^{-3}
Initial Vacancy Loop Number Density, $N_{vI}(0)$	0	m^{-3}
Initial Void Radius, $r_v(0)$	10^{-8}	m
Initial Interstitial Loop Radius, $r_{iI}(0)$	$2.4 \cdot 10^{-8}$	m
Vacancy Loop Formation Radius, $r_{vI}(0)$	$1.5 \cdot 10^{-9}$	m
Gas Atoms per Void, n_g	10	-
Van der Waal's Coefficient, b_v	$8.3 \cdot 10^{-29}$	m^{-3}
Boltzmann's Constant, b	$1.38 \cdot 10^{-23}$	J/K
Vacancy Frequency of Motion, ν_v	10^{13}	s^{-1}
Interstitial Frequency of Motion, ν_i	10^{13}	s^{-1}
Combinatory Number, g	1	-

TABLE 2
Material Properties

	Aluminum	Molybdenum	Units
Burgers vector, b	$2.32 \cdot 10^{-10}$	$2.73 \cdot 10^{-10}$	m
Vacancy formation energy, E_f^V	$1.06 \cdot 10^{-19}$	$4.8 \cdot 10^{-19}$	J
Interstitial formation energy, E_f^i	$5.19 \cdot 10^{-19}$	$1.44 \cdot 10^{-18}$	J
Vacancy migration energy, E_m^V	$9.12 \cdot 10^{-20}$	$2.45 \cdot 10^{-19}$	J
Interstitial migration energy, E_m^i	$1.6 \cdot 10^{-20}$	$1.36 \cdot 10^{-20}$	J
Vacancy diffusion pre-exponential, D_V^0	$4.0 \cdot 10^{-6}$	$3.0 \cdot 10^{-6}$	m^2/s
Interstitial diffusion pre-exponential, D_i^0	$8.0 \cdot 10^{-6}$	$1.6 \cdot 10^{-5}$	m^2/s
Shear modulus, μ	$2.43 \cdot 10^{10}$	$1.15 \cdot 10^{11}$	Pa
Poisson's ratio, ν	0.347	0.32	-
Stacking fault energy, γ_{sf}	0.28	0.574	J/m^2
Surface energy, γ	1.0	2.05	J/m^2
Cascade efficiency, ϵ	0.001	0.001	-
Vacancy dislocation bias factor, Z_V	1.0	1.0	-
Interstitial dislocation bias factor, Z_i	1.08	1.08	-
Damage energy cross section, σ_D	63.0	731.0	b-neV
Threshold energy, E_0	17.0	40.0	eV
800 keV proton energy loss, E^*	$8.0 \cdot 10^{-11}$	$2.4 \cdot 10^{-10}$	J m-f
Current, I	1	0.5	mA
Gaussian size, r_0	$3.1 \cdot 10^{-3}$	$1.18 \cdot 10^{-3}$	m
Sample thickness, l	$2.5 \cdot 10^{-4}$	$2.5 \cdot 10^{-4}$	m

2.1 Numerics

The set of six coupled differential equations is solved using a system routine called SDRIVE, a stiff differential equation solver derived from the same program as used by Ghoniem and Kulcinski (GEAP). The numerical method is based on ideas of Gear [10] and Hindmarsh [11] for the solution of the initial-value problem for systems of ODE's. All calculations were run for 5000 seconds of irradiation.

2.2 Initial Conditions and Material Data

Tables 1 and 2 give the initial conditions and material data used in the calculations. The material data are those used by Ghoniem and Kulcinski except as noted. Four ambient temperatures ($T_0 = 300, 340, 380$ and 400 K) were used for aluminum, and two ($T_0 = 1200$ and 1600 K) for molybdenum. Pulse amplitudes of $T_A = 1, 10, 50$ and 100 K were studied, for seven time constants ($\tau = 0.001, 0.01, 0.1, 1, 10, 100$ and 1000 seconds).

3. RESULTS

Figures 1 and 2 show the void growth calculated after 5000 seconds of steady state irradiation for different ambient temperatures in molybdenum and aluminum, respectively. (The arrows indicate which background temperatures

were used in the series of calculations reported here.) The calculated peak swelling temperature agrees well with experimental data. Molybdenum exhibits void growth for a very large range of temperatures, while aluminum only swells in a narrow temperature window. As expected, at higher temperatures in aluminum ($T > 400$ K) the voids anneal due to large vacancy mobility and thermal emission. (This behavior is also expected in molybdenum, but at much higher temperatures than are considered here.) However, there is also a region of void shrinkage at low temperatures in aluminum ($T < 270$ K). In this regime the vacancies produced during irradiation are quite immobile, and the voids shrink due to absorption of interstitials. The existence of this region plays a dominant role in the effect of temperature pulsing on void growth, as will be demonstrated below.

The temperature was then varied sinusoidally around the selected ambient-temperature values, with various amplitudes and frequencies. Generally, as the amplitude of the temperature oscillation increased, the voids grew more rapidly if the ambient temperature was below the peak swelling temperature; the voids grew more slowly (or shrank faster) if the ambient temperature was above the peak swelling temperature. This behavior is shown in Figure 3 for molybdenum. The effect is more pronounced for $T_0 = 1200$ K.

Figure 1
Void growth as a function of ambient temperature during steady irradiation of molybdenum

which is farther away from the peak swelling plateau than $T_0 = 1600$ K. Figure 4 shows the results of a similar series of calculations for aluminum. For ambient temperatures above peak swelling and for smaller temperatures below peak swelling temperature, the effect of temperature pulsing is that described above (i.e., the material sees an enhanced ambient temperature). The interesting exception occurs when the temperature pulse is large enough that the material spends some time in the low-temperature void-shrinkage regime. If the time spent in that region is small, the void growth is simply retarded. If significant fractions of time are spent there (30-40%), the voids will shrink even when the ambient or time-average temperature is near or above the peak swelling temperature.

This amplitude dependence was seen at all frequencies investigated. Figure 5 shows void growth rate at 500 seconds as a function of the oscillation's time constant for the two background temperatures studied for molybdenum. For both there is a plateau region covering several orders of magnitude in which the void growth seems independent of frequency. The divergence for slow oscillations is readily understandable, since low frequency oscillations allow the system time to react and the void growth rate at 5000 seconds does not correspond to an average growth rate, as shown in Figure 6 for molybdenum and, much more clearly, in Figure 7 for aluminum. For very short pulses

Figure 2
Void growth as a function of ambient temperature during steady irradiation of aluminum

Figure 3
Void growth rate for steady irradiation in molybdenum experiencing sinusoidal temperature pulses of varying amplitude ($\tau = 1$ s)

Figure 4
Void growth rate for steady irradiation in aluminum experiencing sinusoidal temperature pulses of varying amplitude ($\tau = 1$ s)

Figure 5
Void growth rate in molybdenum for steady irradiation with temperature given by $T(t) = T_0 + T_A \sin(t/\tau)$

void growth seems very inhibited, but the reason why is not yet understood.

4. CONCLUSIONS

The fully dynamic rate theory has been used to study the behavior of both aluminum and molybdenum under steady irradiation and sinusoidal temperature fluctuations. Several important conclusions can be drawn:

(1) The calculations correctly predict the qualitative void behavior for steady-state constant temperature irradiation, and the value of the peak swelling temperature. In addition to swelling and annealing, a regime is seen in which voids shrink at low temperatures due to interstitial absorption and vacancy immobility.

(2) If temperature pulses are such that the material temperature always remains above the temperature at which swelling starts, the material always acts as though it sees an enhanced ambient temperature. Void growth is enhanced if the ambient temperature is below the peak swelling temperature and, conversely, void growth is inhibited when the ambient temperature is above the peak swelling temperature.

(3) When the temperature oscillations are sufficient for the material to spend some time in the low-temperature void-shrinkage regime, void growth is first reduced and then replaced by increasing shrinkage as more time proportionately is spent in that region.

Figure 6
Void growth as a function of time for steady irradiation of molybdenum in which $T(t) = 1200 + 10 \sin(t/\tau)$

Figure 7

Void growth as a function of time for steady irradiation of aluminum in which $T(t) = 400 + 10 \sin(t/\tau)$

(4) There is no dependence of average void growth on frequency for many orders of magnitude of temperature oscillations. However, for very short pulses whose characteristic time is similar to the reaction times of the system, void growth seems to be very inhibited.

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